

IV. ENVIRONMENTAL DATA

Dust Measurement

To evaluate either the hazard to health from exposure to dust or the effectiveness of dust control measures, one must have a method or methods for the evaluation of the dustiness. Ideally the methods employed should be as closely related to the health hazard as possible. When determining exposure to dusts containing free silica, in addition to determining the percentage of free silica the method should measure that portion of the dust causing silicosis, ie, that dust which penetrates and is retained in the pulmonary, nonciliated regions of the lungs.

Through the years, many collection methods have been used in the determination of dustiness, and these methods have been reviewed by a number of authors. [78-83] Because these reviews are comprehensive, only the basic principle of the major methods will be briefly discussed here.

(a) Count procedures: The concern of industrial hygienists over the years has been to measure that fraction of a dust that can cause pneumoconiosis. Since it has been recognized that only dust particles smaller than approximately $5 \mu\text{m}$ in aerodynamic diameter are deposited and retained in the lung, methods were sought to measure concentrations of this dust. [83,84] Microscopic counting of dust collected by impingement has long been used for this purpose. Dust counting as an index of dust concentration and consequently of workers' exposure has been used in South Africa by Kitto [85] using the konimeter and in Australia by Owens [86] with a jet dust sampler. In the United Kingdom, thermal precipitation has been frequently used for dust collection [84] while in the United States

the Greenburg-Smith and midget impingers have been commonly employed. [87-89] In these investigations, the lower limit of dust size included in counts was determined by procedure or was implicit in the counting procedure employed. Where a 10X (16 mm) objective lens was used with light-field counting, as in impinger counts, the usual lower limit of diameter of particles seen was approximately 1.0 μm . Others have used dark-field illumination with which it is possible to see particles as small as 0.1 μm diameter. [80]

Because of differences in sampling techniques and instruments used, comparisons of dust concentration with silicosis prevalence in different parts of the world is difficult. This points to the fact that if dust concentrations are measured by count procedure, the procedure employed should follow a standardized method to minimize differences. Such standard methods for impinger sampling and dust counting have been published. [89-91]

Because the prevalence of silicosis in the 1920's and earlier was severe, more effort was devoted to improving dust conditions than to refining and developing methods of dust sampling and measurement. [17,24-29] Although counting methods are inefficient and give variable results, they clearly showed the effectiveness of dust control measures. [25-27,33,44] Later, with efforts to further reduce silicosis, researchers also turned to improving dust measurement methods. [37,79,82,83]

(b) "Total" mass concentration methods: The simplest method of measuring dust concentrations is to determine the total weight of dust collected in a given volume of air. The "total" mass, however, is determined to a considerable extent by the large dust particles, which

cannot penetrate to the pulmonary spaces to cause silicosis. The proportion of dust small enough to penetrate to the pulmonary spaces ("respirable" dust) is extremely variable, ranging in industrial dust clouds from as little as 5% to more than 50% by weight. [80,84]

Thus, the "total" dust concentration by weight is not a reliable index of "respirable" dust concentrations or an index of a silicosis hazard.

(c) Respirable mass size-selective measurement (personal sampling): For evaluation of a silicosis hazard, the method now generally preferred is personal (breathing zone) respirable mass sampling. [78] Dust collection devices now available for this method of sampling also provide a means for a size-frequency analysis of the collected dust. A traditional method for such an analysis has been to collect a sample on a membrane filter and examine it by high-powered optical microscopy (about 1000X), supplemented, perhaps, by electron microscopy, as described by McKee and Fulwiler. [92] Present-day instrumentation permits collection of a dust in such a manner that the sample is size-separated by the design and flow characteristics of the sampling device. Such equipment includes impactors, centrifugal and gravitational separators, and a range of miniature cyclones. [84,92-94] In addition to particle-size separation, these instruments are also capable of collecting a quantity of dust sufficient for an analysis for free silica content of the dust as recommended in Appendix II.

Respirable mass samples are preferably taken over a full 10-hour shift. However, multiple, shorter period (2-4 hour) samples may be collected over an individual's full-shift exposure period, the samples

pooled for analytical purposes, and the average respirable mass concentration of free silica calculated on a full-shift basis. The recommended equipment and the method for collection of dust containing free silica are presented in Appendix I.

Technical Feasibility of Attainment of Standards

(a) Metal mines: Although there is a lack of published information on current dust levels in metal mines, data from Flinn et al [26] substantiate the existence and capability of engineering controls and technology for reducing metal mine dust levels to comply with the recommended standard.

(b) Foundries: In a study to compare impinger counts (mppcf) with results obtained by respirable mass (mg/cu m) sampling for dust containing quartz, Ayer et al [78] compared respirable mass and impinger measurements in a number of Michigan foundries. They found that, in general, foundries that could meet the American Conference of Governmental Industrial Hygienists (ACGIH) Threshold Limit Values (TLV) [95] for free silica by impinger count could meet the ACGIH TLV for free silica by respirable-mass measurement. If reduction of respirable free silica levels in foundries is necessary, the technical means for reducing the dust concentrations to meet the required limit are available.

(c) Ceramics industries: Recent data on free silica concentrations in American ceramic factories are not available. The British Ceramic Research Association [96] has in recent years taken a great number of samples, most of which were personal respirable mass dust samples. As a trade association, this group does not ordinarily publish

results of dust measurements. They seem confident, however, that dust standards somewhat stricter than present ACGIH TLVs could be maintained.

(d) Crushing, grinding, and mixing of minerals containing free silica: Although few workers are employed in individual crushing, grinding, and mixing operations in a given plant, overexposure of workers is common. [44] Existing techniques of enclosure, local and general exhaust ventilation, wetting, and the use of respirators are adequate to reduce to acceptable levels the workers' exposure to dust.

Engineering Control

(a) Foundries: Ventilation designs to control free silica exposures in specific foundry operations are given in the ACGIH Industrial Ventilation manual. [97]

(b) Ceramic Industries: Research and successful application of ventilation, blowing and exhausting, and dilution ventilation have been carried out by the British Ceramic Research Association [98] and have undoubtedly reduced dust levels in this industry in the British Isles. The application of these methods as well as the control procedures outlined in the Industrial Ventilation manual [97] appear to be sufficient to control dust levels in American ceramic plants to the recommended level.

(c) Crushing, grinding, screening, etc: Principles for control of dust from crushers and similar devices and for material handling are given in the Industrial Ventilation manual. [97]

(d) Abrasive blasting: Because of the severe silicosis hazard associated with abrasive blasting with silica sand and the extreme difficulty in controlling the hazards associated with its use in abrasive

blasting, it is recommended that silica sand or other substances containing more than 1% free silica be prohibited as abrasive blasting materials.

[99,100]

V. DEVELOPMENT OF THE STANDARD

Basis for Previous Standards

In the United States, as elsewhere in the world, there are so many dusty trades in which the extent and nature of dust exposure is so varied that the results in one industry are not always comparable to those in another. Table XI-1 emphasizes this variability and shows the concentrations of free silica-bearing dusts that had been accepted as permissible prior to 1940 for the particular industries in the localities indicated. [33]

Hygienic exposure values for dust containing free silica have been based on the quantitative concept that the magnitude of the toxicity is proportional to the concentration of free silica in the dust. When this magnitude of toxicity is represented by an exposure limit, then the limit is inversely proportional to the percentage of free silica in the dust and can be expressed in mppcf as derived from a particle count of the dust-laden environment and a general particle count formula of:

$$\text{Threshold limit} = \frac{K}{\%SiO_2} \text{ mppcf}$$

One of the first recommended "upper limits" for quartz-bearing industrial dusts was that suggested by Russell [33] for the Vermont granite industry based upon studies in that industry. A limit of 10 mppcf for dust containing 25-35% quartz was recommended.

Threshold Limit Values (TLVs) for Chemical Substances and Physical Agents in the Workroom Environment with Intended Changes is a guide adopted by the American Conference of Governmental Industrial Hygienists (ACGIH) for use in the control of occupational health hazards. The value for quartz first published in 1946 [101,102] was originally called a maximum allowable concentration (MAC) value and followed the pattern suggested by the particle count formula given above. However, only three ranges of free silica (quartz) content were considered as indicated below.

<u>Range of SiO₂, %</u>	<u>MAC - mppcf</u>
Silica--High (above 50% free SiO ₂)	5
Silica--Medium (5-50% free SiO ₂)	20
Silica--Low (below 5% free SiO ₂)	50

Review of the early studies of the Public Health Service [17,25,26,29,34] and others [18,32] suggested that the results of the engineering and medical studies were reasonably consistent with values calculated from the count formula using a factor, designated K, equal to 250, and by adding a constant 5 to the percentage of free silica in the denominator. This formula was published by ACGIH in 1962. [103]

$$TLV = \frac{250}{\%SiO_2 + 5} \text{ mppcf}$$

To make the TLV consistent with a 1970 revision of the TLV for nuisance dusts, the numerator K was raised to 300 and the constant 5 raised to 10 in the denominator. [104]

Prior to the 1970 revision of the count formula, a respirable dust concentration formula utilizing respirable mass measurements of dust was introduced [95]:

$$\text{TLV} = \frac{10}{\% \text{ respirable free silica} + 2} \text{ mg/cu m}$$

The formula was based upon the collection of dust by size-selective sampling devices. [82] Such instruments collect a fraction of dust which is capable of penetrating to the gas-exchange portion of the lung where long-term retention of dusts occurs. The concentration of airborne free silica in this size fraction should relate more closely to the degree of health hazard. As with the count formula, a constant was added to the denominator to prevent excessively high respirable dust concentrations when the fraction of free silica in the dust is low. The constant of "2" limits the concentration of respirable dust with less than 1% free silica to 5 mg/cu m.

In addition to quartz, other forms of free silica have been assigned a specific TLV based on experimental or human industrial experience data that indicated a need for individual identification.

Cristobalite (above 5%) was originally listed in 1960 [105] with a TLV of 5 mppcf based on studies in the diatomite industry by Cooper and Cralley [44], Smart and Anderson [48], analogy with the TLV for "silica",

[102] and experimental studies in animals by Wagner et al. [51] In 1968 [95] the TLV was reduced to one-half the value obtained from either the count or mass formula for quartz based upon a review of existing documentation and information to the TLV committee [106] in a personal communication by Smart. This information suggested that the limit of 5 mppcf for cristobalite did not possess a sufficient safety factor for the prevention of pneumoconiosis. Tridymite was likewise assigned one-half the quartz value based upon animal toxicity data developed by King et al [49] in which tridymite was found to be the more active form of free silica studied when its dust was administered by intratracheal injection into the lungs of rats. Analogy was also made with cristobalite.

Although insufficient industrial experience was available to indicate the degree of hazard presented by fused silica dust, the same limit as that required by the quartz formulae was adopted in 1969. [107] Intratracheal injection studies with rats by King et al [49] found fused silica considerably less active than quartz.

Tripoli and silica flour were added to the TLV list in 1972 [108] with the recommendation that the standard for these materials be derived using the respirable mass formula for quartz. Documentation for inclusion of tripoli on the list came from the study of McCord et al [109] who induced tissue proliferation by direct intraperitoneal implantation of tripoli dust in rats and guinea pigs similar to that produced by quartz. Silica flour was included on the list based upon data of King and co-workers [49] and Hatch and Kindsvatter [110] who considered silica flour, because of its fine particle size, to have a significant fibrogenic potential.

The 1968 ACGIH recommended TLVs for quartz have been adopted by the US Department of Labor under the Walsh-Healey Public Contracts Act regulations (41 CFR 50. 204). The TLVs have also been adopted by the US Department of Interior under the Metal and Nonmetallic Health and Safety Act (Sec 6, 80 stat 774; 30 USC 725).

The Federal Coal Mine Health and Safety Act of 1969 (PL 91-173) provides that the Secretary of Health, Education, and Welfare prescribe a formula for determining the applicable standard for coal mines where quartz amounts to more than 5%. Such a formula has been published using 0.1 mg/cu m of respirable quartz as a basis. [30 CFR Part 70.101 published in Federal Register, vol 36, page 4941, dated March 16, 1971 and 30 CFR Part 71.100 published in Federal Register, vol 37, page 6368, dated March 28, 1972] The limit thus becomes: mg/cu m respirable dust = 10/% quartz. The 1968 TLV for free silica and the statutory limit for quartz at 5% and 2 mg/cu m of respirable dust are the principal bases for the quartz limit for coal mines. Thus the allowable level of airborne quartz in coal mines is 100 µg/cu m, twice the limit recommended in this criteria document.

Because methods employed by different countries for assessment and reporting of dust concentrations in the workplace vary considerably, comparison of standards recommended by various countries for exposure to dust containing free silica cannot be made with certainty. Examples of standards for free silica adopted by several countries other than the US follow.

The Federal Republic of Germany has adopted a MAK value of 0.15 mg/cu m for quartz, including cristobalite and tridymite. According to Schutz, [111] this standard is based upon a comparison of different local

and foreign standards, results of silicosis statistics, mean dust levels in several industries, and calculations based upon the amount of dust which could be retained in the lungs of exposed workers.

The Swedish values for quartz use a gravimetric determination of the total amount of dust per cubic meter of air. [112] This quantity is related to the free silica content if this mineral exceeds 2.5%. By a formula based on dust quantity and percentage of free silica, a dust index is derived which relates to a given exposure limit. Values exceeding 1.0 represent a silicosis risk. If the free silica is less than 2.5%, a standard of 15 mg/cu m is used. If dust contains large amounts of cristobalite or an unusually great proportion of respirable particles, a lower value is applied.

DeGuedre, [81] in a review of methods adopted by different authorities for assessing the hazard relating to exposures to dusts in mines, included the following standards for France and the USSR on a list of criteria without identifying the basis for their adoption.

France has required a dust index for each workplace since 1956. The index is derived from a formula utilizing the number of dust particles per cu cm below 5 μ m, the percentage content by weight of free silica as determined by X-ray diffraction of dust below 5 μ m, and a constant dependent upon the sampling and examination methods adopted. The dust index, related to the silicosis risk, determines the frequency of medical examinations. Dust concentrations with an index of 5 or less are considered satisfactory; between 5 and 6, doubtful; and those above 6, dangerous.

The USSR expresses a standard in which "maximum permissible concentrations are as weights of fine dust, probably under 5 microns."

Based upon the percent free silica, the following concentrations are permitted. [81]

<u>Mineral and Organic Dust</u>	<u>Maximum Permissible Concentration mg/cu m</u>
Over 70% crystalline silica	1
10-70% free silica	2
Silicate dust with 10% free silica	4
Other mineral dust with 10% free silica	5
Minerals and mixtures with no silica	6
Coals with more than 10% free silica	2
Coals with less than 10% free silica	4
Coals with no silica	10

The present federal standard for free silica exposure is an 8 hour time-weighted average based upon the 1968 ACGIH TLV formulas of $250/\%SiO_2 + 5 = \text{mppcf}$ or $10 \text{ mg/cu m}/\%SiO_2 + 2$ for respirable quartz. One-half this amount has been established as the limit for cristobalite and tridymite. [29 CFR Part 1910.93 published in the Federal Register, volume 39, page 23543, dated June 27, 1974]

Basis for Recommended Environmental Standard

The literature contains many publications on exposures to dusts containing free silica. Unfortunately, data necessary for development and recommendation of a standard to protect the health of workers against the harmful effects of exposure to such a potent pneumoconiosis-producing material are seldom contained in the published reports. [18,47] Epidemiologic studies too frequently have not included environmental data.

or such data, if available, have related only to the then present conditions with no correlation with past exposures. In addition, reevaluation studies of a given industry on the state of the worker's health resulting from continued exposure to free silica-bearing dusts have not generally been made. Refinements in the technology of sampling and analysis of dusts and of methods for monitoring biological response will make possible a more precise and valid evaluation of the effects of exposure to dusts which may cause silicosis.

A review of the data from epidemiologic studies of workers in metal mines [25,26] and in foundries [27] reveal that the medical data are reasonably consistent with impinger-count dust concentration data. In these studies the prevalence of silicosis was reduced significantly in the work environments where dust levels were controlled at or below 10 mppcf. Additional data on the effects of exposure of workers at dust levels below 10 mppcf can be found from the studies in the pottery industry, [29] the silica brick industry, [30-32] and the granite industry. [17,33,36-39]

In pottery workers Flinn et al [29] found that at dust levels of less than 8 mppcf there was an increasing prevalence of silicosis with increasing length of exposure at dust levels between 4.0-7.9 mppcf. The prevalence of silicosis ranged from 0.3% among workers exposed for less than 10 years to 85% among workers with over 40 years of exposure at this level (see Table III-2). Two cases of early silicosis were observed in 798 workers exposed for 10-29 years at less than 4 mppcf. However, these cases could have received higher exposure at some previous work period and thus one cannot say with certainty that they occurred as a result of the lower exposure.

While the study of Fulton and co-workers [32] of the silica brick industry showed a significant prevalence of silicosis of 52% in 1,035 employees examined, interpretation of these data must take into account the absence of free silica dust exposure data for several years preceding the survey. Such information is considered essential for an accurate evaluation of the reported health effects due to the inhaled dust. This is particularly true in light of the 42% prevalence of silicosis found in workers exposed at an average concentration of 0-9.9 mppcf as determined at the time of the 1939 study. In all probability dust exposures significantly higher than 10 mppcf were experienced in previous years by the workers in these same operations and could have been responsible in part for the recognized cases of silicosis. The reported 22% (14 of 65) incidence of silicosis in workers examined where average dust exposures were 2-3.9 mppcf would tend to support the conclusion that higher dust exposures contributed to the prevalence of silicosis. Review of the literature on silicosis revealed no other report with unequivocal diagnosis of silicosis based on dust exposures at levels between 2-4 mppcf.

The study of Fulton et al suggests that cristobalite and tridymite have a capacity greater than that of quartz to induce silicosis. An average length of exposure of 17.9 years was required to produce stage 2 silicosis among the green-brick workers whose exposure was restricted to that form of dust containing 88% quartz. Men in the burned-brick department were found to have stage 3 silicosis after an exposure of like duration to burned-brick dust of 80% cristobalite and tridymite. Average dust concentrations were essentially the same, 15.9 and 16.9 mppcf, respectively. Animal studies confirm the greater activity of cristobalite

and tridymite. [49,53] Interpretation of available data from the silica brick study [32] suggests an exposure limit for free silica dust below 10 mppcf. However, the enhanced biological response resulting from exposure to the mixed cristobalite-tridymite dust would suggest a limit closer to 5 mppcf as being more appropriate for an exposure level at which silicosis should not occur. However, a greater toxicity of cristobalite and tridymite than of quartz, if expressed as respirable mass rather than as particle count, does not necessarily follow from the epidemiologic studies based on impinger count.

Surveys of the diatomite industry in California [44-46] associate a 9% incidence of silicosis in workers exposed to dusts from calcined diatomite containing up to 61% cristobalite in the parent material and up to 32% in airborne dust. Reduction of dust concentrations to a point where more than 84% of the samples counted were routinely below 5 mppcf reduced the incidence of silicosis to zero in workers whose employment began after dust control measures were instituted. The 5 mppcf level was suggested as the maximum exposure concentration for the industry. [44] Subsequent studies, [45,46] the most recent being conducted 16 years after the initial survey, appear to confirm the validity of the 5 mppcf level for dusts containing cristobalite. Again, no evidence of silicosis was reported for individuals employed since initiation of the dust control program.

It is from the Vermont granite industry that the most extensive and complete environmental and medical data are available for establishing a recommended environmental limit for exposure to free silica. These data have been accumulated over approximately a 50-year period extending from the 1924 study of Russell et al [17] to that of Theriault and co-workers

[37-39] in 1969-72. With the exception of the reports by Theriault et al, all occupational environment dust exposures were determined by microscopic counts of impinger-collected dust samples. The derived air dust concentrations in mppcf and the associated health effects provide the major portion of the material used as the bases for the present exposure limits (TLV's) for quartz and other free silica polymorphs [95,104,107,108] in addition to the 10 mppcf granite dust exposure limit in use in that industry since 1937. [33-35]

The studies of Theriault and co-workers [37-39] of Vermont granite workers are important from several standpoints, among them their use of size-selective respirable mass sampling, coupled with gravimetric determinations of dust concentrations, instead of impinger counts, as in past studies.

These investigators [38-39] also based interpretations of granite dust toxicity on pulmonary function tests as well as on X-ray evidence. While the observed average decrement in function may not have clinical significance, it appears to presage radiographically evident changes, and thus could be a more sensitive index of effects in a group of workers, whether or not it would be sufficiently sensitive for diagnosis of disease in an individual. Yet, these studies do not demonstrate a safe concentration of silica. The authors found that 50% of the workers had radiographic evidence of silicosis at 46 dust-years (ie 46 years of exposure at a dust level equivalent to about 50 $\mu\text{g}/\text{cu m}$ of free silica) and functional evidence at over 32 dust-years. But the curves drawn to fit their data suggest a significant incidence of silicosis at 0 dust-years. [39] Based on a plot of radiographically evident silicosis against dust-

years, 30% of the working population had silicosis with no exposure. Of course, this undoubtedly represents imperfections in their data or in available methods of analysis of their data, but it is not evident how to use inferences from these analyses in deriving an environmental limit. One can speculate that their environmental samples were unrepresentative of the years of exposure preceding the sampling, which surely contributed to the development of silicosis; the authors attempted to make some estimate of the extent to which past exposures might have been higher than indicated by their more recent atmospheric sampling and, though their estimates seem reasonable, they could have been in error. Another curious observation was that the silica content of the dust sampled (9%) was much lower than previous analyses had demonstrated; they suggested that this was due to process changes which caused a dilution of the silica content of the total dust. It is believed important that these studies be confirmed in the granite sheds and be extended to other operations producing airborne silica, as a likely prerequisite to further refinement of an occupational health standard for free silica.

Russell et al [17] studied 972 granite shed workers, dividing them into 4 exposure groups according to average dustiness: 37-60, 27-44, 20, and 3-9 mppcf. The group with the highest dust exposure showed the unmistakable indication of the seriousness of the hazard of exposure to granite dust by development of early silicosis in 40% of workers after two years and 100% after 4 years of exposure. The development of silicosis in the remaining groups appeared to be proportional to the dust exposure. An experience similar to the highest exposure occurred at the second highest

exposure (27-44 mppcf) where early stages of silicosis appeared after 4 years of exposure and more advanced stages developed by the 7th year.

In the group exposed at an average of 20 mppcf there was little indication of severe effects upon the health of the workers. However, the authors concluded that one would hesitate to state positively that no harm would come to persons exposed for many years to a concentration under 20 mppcf. In the case of the lowest exposure group where the average dust concentration was 6 mppcf (range 3-9 mppcf), there was no indication of any untoward effects of dust exposure on workers.

From the above, the authors [17] interpreted that average dust exposure for the 2 highest exposure groups was clearly harmful to workers. They concluded that, even though harmful effects were found, a safe limit of dust exposure apparently lies somewhere between 10 and 20 mppcf.

In a restudy of the granite workers [33] Russell revised his original estimate [17] of 10-20 mppcf as the desirable limit for granite dust exposures. Rather than basing his new recommendation upon data from the 20 mppcf average granite dust exposure group, which still carried "some question as to the harmful effect" of the dust exposures at that average concentration, Russell apparently used the progression of silicosis from the milder forms to the more severe forms; he also used the further complication of tuberculosis in the highest average dust exposure group (27-44 mppcf) as the basis for his new limit of about 10 mppcf.

Following this study and enforcement of the 10-mppcf limit by the Vermont Department of Health, dust control progressed in the granite sheds so that by the time of the study by Hosey et al [34] few exposures in the granite sheds studied exceeded 5 mppcf. The effectiveness of the control

measures was evidenced by the absence of new cases of silicosis, with the exception of one doubtful case, in men starting work in the granite sheds after 1937. Furthermore, chest roentgenographic surveys of granite workers showed a reduction in the prevalence of silicosis from 45% in 1937 to 15% in 1956.

Confirmation of the safety of the limit of 5 mppcf was reported by Ashe and Bergstrom [35] in 1964. Their study, 26 years after dust control began, likewise found no cases of silicosis in workers employed after the start of dust control. Environmental data also indicated a probable greater margin of safety for dust exposures at the time of the study; average concentrations were 3 mppcf.

Based on the impinger-count dust concentration data and the reported absence of identifiable dose-response effects, the granite shed studies [33-35] indicate that a limit of 5 mppcf has, up to this time, been an effective control for the prevention of silicosis in men exposed to granite dust of 25-35% quartz.

As had most of the investigators before them, Hosey et al [34] and Ashe and Bergstrom [35] concluded that careful surveillance of the work environment and of the worker's health was needed to determine the ultimate efficacy of dust control in the granite sheds industry.

From the above studies in Vermont granite sheds, a safe level for silica can be interpreted as 5 mppcf. Because of variations in types, size, and density of particles in other industries, it is not clear that the same limit, in terms of number of particles, will properly describe safe exposures in these other industries producing airborne free silica. But on the basis that 5 mppcf is equivalent, in Vermont granite sheds, to

50 $\mu\text{g}/\text{cu m}$, (see discussion below) it seems appropriate to apply this limit, in terms of respirable mass, to other operations producing dusts containing free silica. Thus, an environmental limit of 50 μg of respirable free silica/cu m is recommended.

Reno et al [113] and Sutton and Reno [114] compared impinger-count measurements with size-selective mass concentrations from granite shed worker environments in an attempt to establish a relationship between these methods of sampling free silica. They concluded that 10 mppcf of granite dust (containing approximately 25-35% free silica) was equivalent to 100 $\mu\text{g}/\text{cu m}$ of free silica. Their work has been reviewed and evaluated by Ayer and his associates. [78,79] On the basis of their review, Ayer et al supported the conclusion of Reno and Sutton and their collaborators, ie, that 10 mppcf of total granite dust is approximately equivalent to 100 $\mu\text{g}/\text{cu m}$ of respirable free silica. Theriault and associates [37] came to a slightly different conclusion, viz, that 10 mppcf is equivalent to about 80 $\mu\text{g}/\text{cu m}$, but they presented no data or argument supporting the conclusion. Thus, a safe level of silica of 5 mppcf for the granite workers indicates a level of 50 $\mu\text{g}/\text{cu m}$ in terms of respirable free silica.

A review of data from other industries [26,27,29,32,44] does not reveal any significant difference in the degree of toxicity of free silica in the form of quartz to which workers are exposed as compared with that in the granite industry. They do reveal, however, that the 10 mppcf standard has not been entirely adequate for protection of workers in those industries, a condition which has been suggested by the data from the granite industry. Consequently, the recommended standard of 50 $\mu\text{g}/\text{cu m}$ is considered applicable to all work environments where exposure to the quartz

form of free silica may occur. It is recommended that the studies in the granite industry be confirmed and that similar studies be undertaken in other industries to determine more precisely the significance of exposure to free silica in those industries so that alternate recommendations can be made should they be indicated.

The epidemiologic studies of Fulton et al [32] and Cooper and Cralley [44] have suggested that cristobalite and tridymite are more active than quartz in producing fibrotic change in lung tissue. King et al [49] and Gardner [53] have confirmed this in animal studies. In addition, experimental evidence indicates that microcrystalline free silica, because of its extremely fine particle size, may have a greater potential for inducing fibrotic change. [56,110] Because of these factors, it has been recommended [95] that a standard for these forms of free silica be one-half that recommended for quartz. Regrettably, there are no studies which relate mass respirable quantities of cristobalite, tridymite, or microcrystalline free silica to a prevalence of silicosis in an exposed population. However, the epidemiologic studies cited [32,44] above and follow-up studies in the diatomite processing industry [45,46] have indicated that if exposure levels of cristobalite and tridymite are kept below 5 mppcf no cases of silicosis are likely to develop in an exposed worker population. Similar data for microcrystalline free silica are lacking.

While the respirable mass concentration of 50 $\mu\text{g}/\text{cu m}$ cannot be shown at this time to be equivalent to the 5-mppcf particle count concentration in operations other than granite work, it is believed that a free silica concentration of 50 $\mu\text{g}/\text{cu m}$ in air is sufficiently low to

protect workers exposed to cristobalite, tridymite, or microcrystalline free silica against the development of silicosis, thus no separate standard for these forms of free silica is recommended at this time. Further research is needed to validate inferences about the safety of the 50 $\mu\text{g}/\text{cu m}$ limit for other forms of free silica; meanwhile, it is recommended that the limit of 50 $\mu\text{g}/\text{cu m}$ should apply to any form of free silica.

Despite the questions raised above about the studies of Theriault and associates, [37-39] their approach seems clearly superior to that of past studies. Respirable mass sampling of worker populations should give a lower variance in results and should show more clearly whether other dusts may potentiate or antagonize silica toxicity (Theriault et al suggested that other components of granite dust slightly increased the toxicity of silica). Perhaps more importantly, correlation of effects judged by X-ray evidence and by pulmonary function tests would be expected to demonstrate the superiority of pulmonary function tests as an early indicator of silicosis (and for this reason, tests of pulmonary function are recommended for routine medical monitoring of silica workers in this recommended standard), and such testing should be included in the design of further research.

It is recognized that many workers are exposed to small amounts of free silica or are working in situations where, regardless of amounts used, there is only negligible contact with the material. Under these conditions it would not be necessary to comply with many of the provisions of this recommended standard, which has been prepared primarily to protect workers' health under more hazardous circumstances. Concern for workers' health requires that protective measures be instituted below the enforceable limit

to ensure that exposures stay below that limit. For these reasons "exposure to free silica" has been defined as exposure above half of the environmental limit, thereby delineating those work situations which do not require the expenditure of health resources for environmental and medical monitoring and associated recordkeeping. This level has been chosen on the basis of professional judgment rather than on quantitative data that delineate nonhazardous areas from areas in which a hazard may exist.

The length of time necessary for silicosis to develop when workers are exposed to relatively low levels of free silica makes it necessary to retain medical and environmental records for extended periods of time for effective evaluation of control measures. The time of retention of these records as they relate to workers exposed to free silica should be at least 30 years following termination of employment.

Subsequent to the completion, review, and approval of this document, a summary of new information was furnished NIOSH (personal communication, H. Ohman, Vasteras, Sweden, September 1974). At exposure levels, expressed in increments of 10 $\mu\text{g}/\text{cu m}$, from 10 to 280 $\mu\text{g}/\text{cu m}$ of respirable free silica, none of the groups exposed at concentrations up to 50 $\mu\text{g}/\text{cu m}$ had radiographic evidence of silicosis, but at all higher levels there was at least one case of silicosis, the percentage affected increasing with concentration level. Exposures were calculated on the basis of 40-year exposures as constituting a working lifetime. If review and analysis of the data and methods, information not now available, supports the inferences based on the summary, the study would offer additional evidence for the environmental limit of 50 $\mu\text{g}/\text{cu m}$, in this case from foundry operations.

Basis for Recommended Sampling Method

"Impinger sampling combined with its microscopic counting method has served well in the past as a tool in reducing exposures to dusts which give rise to pneumoconiosis". [115] However, in spite of its success as a monitoring method, the impinger is deficient in most of the factors which are desirable for evaluating a dust standard. [79] Results obtained with the impinger are not closely related to the health hazard when dust is in the form of agglomerates, as is the case of most redispersed dust. Many dust particles are sufficiently large so that when they are inhaled they are removed by the upper respiratory tract. Thus they never reach the pulmonary spaces where tissue change can occur, yet they are collected and counted by the impinger method and are considered in the total count. Where virtually all dust is in the form of discrete, respirable-size particles, the counts may be very much lower, even though the hazard is far greater than for a dust of greater size. [79] In addition, careful training of dust counters is required before their counts approach the average value of experienced counters. The cost of the determination of an average exposure is high. Any one impinger sample usually measures only 10-30 minutes of exposure and at least 5 samples are required to determine an exposure with any degree of confidence. [82] It is evident that the impinger method falls short of the ideal with regard to relevance to health hazard, simplicity, reproducibility, and unit cost. [79]

The Johannesburg Conference on Pneumoconiosis of 1959 [116] recommended that "measurements of dust in pneumoconiosis studies should relate to the 'respirable fraction' of the dust cloud...". During its 1968

annual meeting, the ACGIH accepted a report of the TLV Committee which recommended adoption of a quartz TLV for respirable dust in mg/cu m. [106]

The use of size-selective sampling of respirable dusts as a means of evaluating inhalation hazards has been reviewed by Hatch and Gross, [84] by Morrow, [83] the AIHA-ACGIH Aerosol Technology Committee, [82,117] and Ayer et al, [78] and all have well stated the advantages to be gained by using this method of sampling. These include: the ability to sample over a full shift or a major fraction thereof; automatic compensation by the size-selective device for shape, density and degree of agglomeration of dust; the ability to use personal samplers and obtain truer "breathing zone" exposure; the greater possibility for standardizing analytical determinations (as contrasted with optical count); and a much lower cost per determination of weighted exposure to dust. The samples can also be used for determination of free silica content, weight concentration, and particle size distribution.

The size-selective, respirable mass collection of dust provides a method and data that can be more closely related to the health hazard associated with the inhalation of free silica particles. The method is simple, reproducible, and relatively inexpensive. The size-selective mass method separates out the large dust particles by an inertial or gravitational method, allowing only those sizes of dust to pass which are capable of penetrating to the pulmonary, nonciliated portion of the lung. The method is ideally suited for collection of the essentially insoluble free silica dusts which exert their damaging effect in the pulmonary area [79] and is the method recommended for collection of dust samples for

evaluation of a silicosis hazard. Detailed procedures for application of the method are given in Appendix I.

Basis for Recommended Analytical Method

Three principal methods are currently used for the qualitative and/or quantitative determination of free silica in workplace dusts. These analytical methods are: the colorimetric chemical procedure, infrared spectrophotometry, and X-ray diffraction.

At present the colorimetric procedure [118-121] is the method most universally used. However, there are two serious drawbacks to this wet chemical method which prevent its recommendation as the method of choice: (1) the analytical results are highly operator-dependent, requiring extreme adherence to a timed, precise protocol due to color instability; (2) the method does not distinguish between the free silica polymorphs--quartz, cristobalite, and tridymite--which at present have different Federal standards for permissible airborne concentrations. [29 CFR Part 1910.93 published in the Federal Register, volume 37, page 22139, dated October 18, 1972]

The infrared procedure [40,122-125] is a relatively new analytical method for free silica which has the potential for the qualitative identification of the free silica polymorphs. [126] The method has been routinely applied for the determination of quartz only. [122,127] Another drawback is the dependence of the analytical results on particle size. [123,125,128] Samples having an average particle size greater than 2 μm have a reduced absorbance at the analytical bands of 12.5 and 12.8 μm . [129]

The X-ray diffraction procedure, [130-132] on the other hand, is specific for the various forms of free silica, [133] including the microcrystalline variants. [134] The method is sensitive, detecting as little as 25 μg of quartz on a silver membrane filter. [135] Moreover, this procedure requires less sample preparation than either the infrared or the colorimetric procedures. [136]

Comparative studies by NIOSH of the three analytical methods utilizing field samples of respirable Georgia granite dust indicate that all three give equivalent percentages of free silica on field samples. The results of analyses of 45 side-by-side granite shed dust samples, [137] collected on three different days, are presented in Table V-1.

TABLE V-1
PERCENTAGE OF FREE SILICA RECOVERED FROM GEORGIA GRANITE DUST
BY THREE DIFFERENT ANALYTICAL METHODS

<u>Analytical Method</u>	<u>Number of samples</u>	<u>Mean % free silica</u>	<u>% Deviation from overall mean</u>
Colorimetric	18	23.6	+0.9
Infrared (512 cm^{-1})	12	24.5	+5.0
X-ray Diffraction	<u>15</u>	<u>22.2</u>	<u>-5.0</u>
	45	23.4 (overall mean)	

None of the methods described above are ideal for analysis of dust samples for free silica under all conditions [137]; but, because of its sensitivity, speed, minimum sample preparation time, ability to identify the polymorphs of free silica, and capabilities for automation, the X-ray diffraction method is recommended as the method of choice for the quantitative and qualitative analysis of dust containing free crystalline silica. Detailed procedures for application of the method are given in Appendix II. When conditions warrant, the colorimetric or infrared spectrophotometry methods may be used. Such conditions may occur: (1) when interfering materials in the sample will decrease the sensitivity of the X-ray diffraction method by blocking the primary diffraction peaks; (2) when more than 1 polymorph of free silica is present which would interfere with the accuracy of the results obtained; (3) when the quantity of the total sample is small or when cristobalite, tridymite, or other polymorphs of free silica are a significant fraction of the sample. The experience of the laboratory performing the analysis and their knowledge of conditions under which the samples being analyzed are collected will, in a large measure, determine which alternate method should be used.

When infrared or colorimetric analytical methods are used, the procedure for these methods [138,139] as given in the NIOSH Manual of Analytical Methods should be followed.

These methods will provide as accurate a means for qualitative and quantitative analysis of free silica in collected respirable dust samples as is presently available.